Liu *et al.* Reply: A main concern raised in the Comments [1,2] is the reliability of the 2.6 Å shoulder peak in the pair distribution function (PDF) [3]. Here we provide more details to further clarify this issue. For obtaining the PDF, we used the truncated Lorch-window method for background subtraction and compared multiple diffraction patterns in different regions of the same sample. The background curve links the central points of each major peak and overlaps with the high-Q experimental data. The signals are sharp in the low-Q range (< 12 Å^{-1}), and also significant in the higher Q range. Figure 1(a) shows the comparison of different truncation ranges, with the enlarged Q range from 12 to 16 Å^{-1} in the framed inset. The three apparent peaks were almost smeared out by the aggressive Lorch-window filtering with a truncation value of 12 Å^{-1}, which would eliminate the shoulder peaks in the Fourier transform in Fig. 1(b). The truncation of 16 Å^{-1}, in comparison, keeps the original signals, leading to and the revelation of the presence of shoulders inthe PDF.

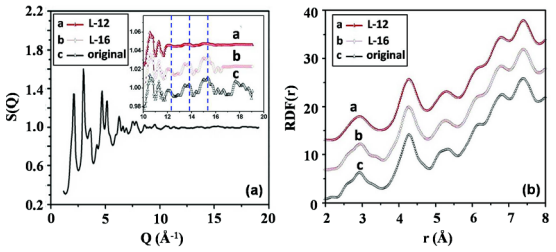

FIG. 1 (color online). (a) Original structure factor $S(Q)$ (black) and truncated $S(Q)$ data. The inset in (a) enlarges the Q range from 10 to 20 Å^{-1}, for different Lorch-window filtering. (b) Corresponding RDF curves. L-12 (red half-solid triangle) represents the data with a truncation range of 12 Å^{-1}, and L-16 (open square) for the truncation range of 16 Å^{-1}.

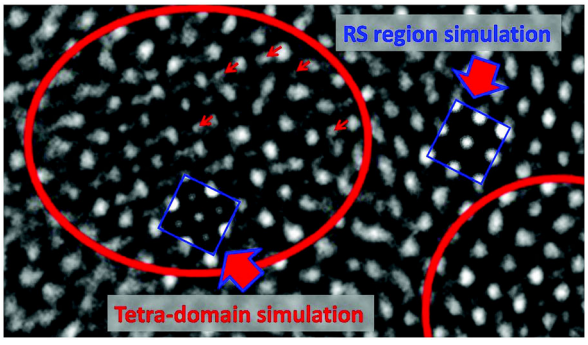

FIG. 2 (color online). Cs-corrected HRTEM image showing VtGC domains (inside red circles) coexisting with RS regions (outside red circles). Some atoms are clearly off-octahedral and close to being tetrahedral (marked using small red arrows). Overlapped on top in blue squares are images simulated using models for the VtGC and RS regions, respectively, under our experimental conditions.

The other question raised is about the TEM image analysis. As shown in Fig. 2, Cs-corrected HRTEM image contains both rocksaltlike (RS) domains and domains with tetrahedral Ge. The two types of domains appear next to one another, thus excluding effects of sample thickness. Systematic image simulations have been performed, for the sample thickness (20–30 nm) and defocus parameters used in the experiment. For the RS structure, no ghost spots appear at tetrahedral sites under all simulation conditions. In contrast, when VtGC domains [3] are added in the simulation model, tetrahedral-Ge spots appear in the overfocus range of 2–8 nm. Representatives of the simulated HREM images (blue squares) are overlapped with the experimental ones in Fig. 2. The direct comparison reveals the presence of atoms in distorted tetrahedral sites, as indicated by small red arrows.

There have also been hints suggesting the existence of off-octahedral Ge atoms, from EXAFS [4], neutron diffraction [5], high resolution transmission electron microscopy [6], and also x-ray fluorescence holography [7].

To reduce energy, the tetrahedral Ge atoms prefer to cluster with shared vacancies, leading to domains [3]. While energetically the RS structure is more favorable [2,3], other structure variations may come in, including those with rather limited metastability, during rapid phase transition. Some of the tetrahedrally bonded Ge in the amorphous GST phase (up to ~30% of all Ge) [8] can conceivably be retained in the first emerging crystalline configuration. While our findings are instructive, we do agree that caution should be exercised before generalizing the findings from a specific TEM sample. Further efforts are needed to firmly conclude on the exact atomic fraction and bonding nature of tetrahedral or off-octahedral Ge.

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